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An Analysis of Surface Production of Sea-Salt Aerosols

C. W. Fairall
BDM/NPS
Monterey, California 93940

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FOREWORD

This report was prepared under Work Order Number 422 of Contract N00014-78-C-0204 in support of the U.S. Naval Postgraduate School research project supported by the Naval Air Systems Command (Air 370) and the Naval Material Command (EO/MET). The work was performed in support of the Environmental Physics Group at NPS under the direction of Professors G. E. Schacher and K. L. Davidson.

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ABSTRACT

The rate of production of sea water droplets by bubble bursting at the ocean surface is virtually impossible to measure directly. Laboratory simulations are also difficult. However, the surface flux can be inferred from measurements of the rate of change of aerosol densities and changes in the height of the marine boundary mixed layer. Data from CEWCOM-78 has been analyzed to produce the aerosol surface flux volume spectrum from 0.5 to 15 μm radius at a wind speed of 8 m/sec. Using this flux spectrum and equilibrium aerosol spectra from JASIN, similar flux spectra are calculated for sind speeds from 5 to 15 m/sec.

A. INTRODUCTION

There are two components to aerosols in the marine boundary layer: 1) continental (background) and 2) locally generated sea-spray droplets. The sea-spray droplets are generated primarily by the bursting of bubbles at the sea surface. The bubbles are produced by biological activity, chemical reaction and breaking waves (white caps). At wind speeds greater than about 3 meters per second, white caps are the primary contributor to the bursting bubbles. Thus, the sea surface is a continuous source of sea-salt aerosols in the marine boundary layer. These surface produced aerosols can be characterized by a surface flux spectrum, $F_s(r)$, which represents the volume of aerosol produced per square centimeter of ocean surface each second as a function of aerosol particle radius. This quantity is a function of wind speed.

The continuous production of sea-salt aerosols is balanced by several removal mechanisms. One obvious mechanism is the loss of particles as they fall back to the surface. This settling under gravity is called "Stokes fallout" and is characterized by the Stokes velocity. The particles are transported vertically by turbulence in the marine boundary layer and maintained at a uniform density throughout the mixed layer. The growth of the height of the layer constitutes another loss mechanism called "entrainment." The final loss mechanism is "rainout," which occurs when the particles become condensation nuclei in the formation of clouds.

Given the surface flux spectrum and a parameterization of the removal mechanisms, one could predict evolutions of the aerosol density spectrum. Unfortunately, the surface flux spectrum is not known. However, we can reverse the process--that is, use parameterizations of the removal processes and evolutions of the aerosol spectrum--to obtain estimates of the surface flux spectrum. This report describes the calculation of $F_s(r)$ from data taken by the NPS Environmental Physics Group during CEWCOM-78 and JASIN.

B. AEROSOL SPECTRUM

The first step to analyzing the evolution of the aerosol spectrum is to remove those variations due to changes in the ambient relative humidity. This is done by transforming each spectrum to a reference relative humidity (RH = 80% or a saturation ratio S = 0.8). We make the usual assumption that the particle radius a saturation S is given by:

$$r = r_0 \; g(s) \tag{1}$$

where r_0 is the radius at S = 0.8 and $g(s) = 0.81 \exp [0.066S/(1.058 - S)]$. If we have a measured aerosol number density spectrum, $n(r)$, then the transformed spectrum at standard saturation, $n'(r_0)$, is:

$$\frac{dn'}{dr_0} = n'(r_0) = n\,(r_0\, g(s)) \;\; g(s) \tag{2}$$

Since we prefer to work with the volume spectrum $V(r) = \frac{4}{3}\pi r^3 n(r)$, the corresponding relationship is:

$$\frac{dV}{dr_0} = V'(r_0) = V(r_0 g(s))/g^2(S) \tag{3}$$

We now must separate the measured spectrum into its two basic components:

1) the background continental aerosol of nonlocal origin, V_c, which is present above and within the marine layer; and

2) the sea-salt aerosol locally generated at the sea surface, V_s, which is well-mixed throughout the marine layer ($Z < h$).

Thus, we assume:

$$V(r) = V_c(r) + V_s(r) \qquad Z < h \tag{4a}$$

$$V(r) = V_c(r) \qquad Z > h \tag{4b}$$

The continental aerosol component is represented with a Junge type distribution,

$$V_c(r) = A/r \tag{5}$$

where A is the continental coefficient. Since there is very little sea-salt volume production for $r_o < 0.3$ μm, we can use the small size range of the spectrum to calculate the continental coefficient ($A' = r_o V'(r_o)$). This is documented in Figure 1 where ensemble averages of volume spectra indicated that $V'(r_o)$ is essentially independent of wind speed at $r_o = 0.1$ μm. At low level wind speeds, the $r_o = 0.1$ μm and $r_o = 0.3$ μm ensemble average spectra are described quite accurately by Equation 5. The claim that A' is an accurate index of continental influence is nicely validated by comparison with atmospheric Radon activity from CEWCOM-78 (Figure 2). Thus, locally generated sea-salt component can be calculated using:

$$V_s'(r_o) = V'(r_o) - A'/r_o \tag{6}$$

The remainder of this report will deal exclusively with the volume spectra transformed to standard humidity, $V_s'(r_o)$. However, to simplify the notation we will drop the prime and sub-zero with the understanding the V_s now refers to the spectrum at RH = 80%, with radius, r, an implied variable.

The evolution of the aerosol spectrum is described by the following equation:

$$h \frac{dVs}{dt} = F_s - (We + Ws)\ Vs \tag{7}$$

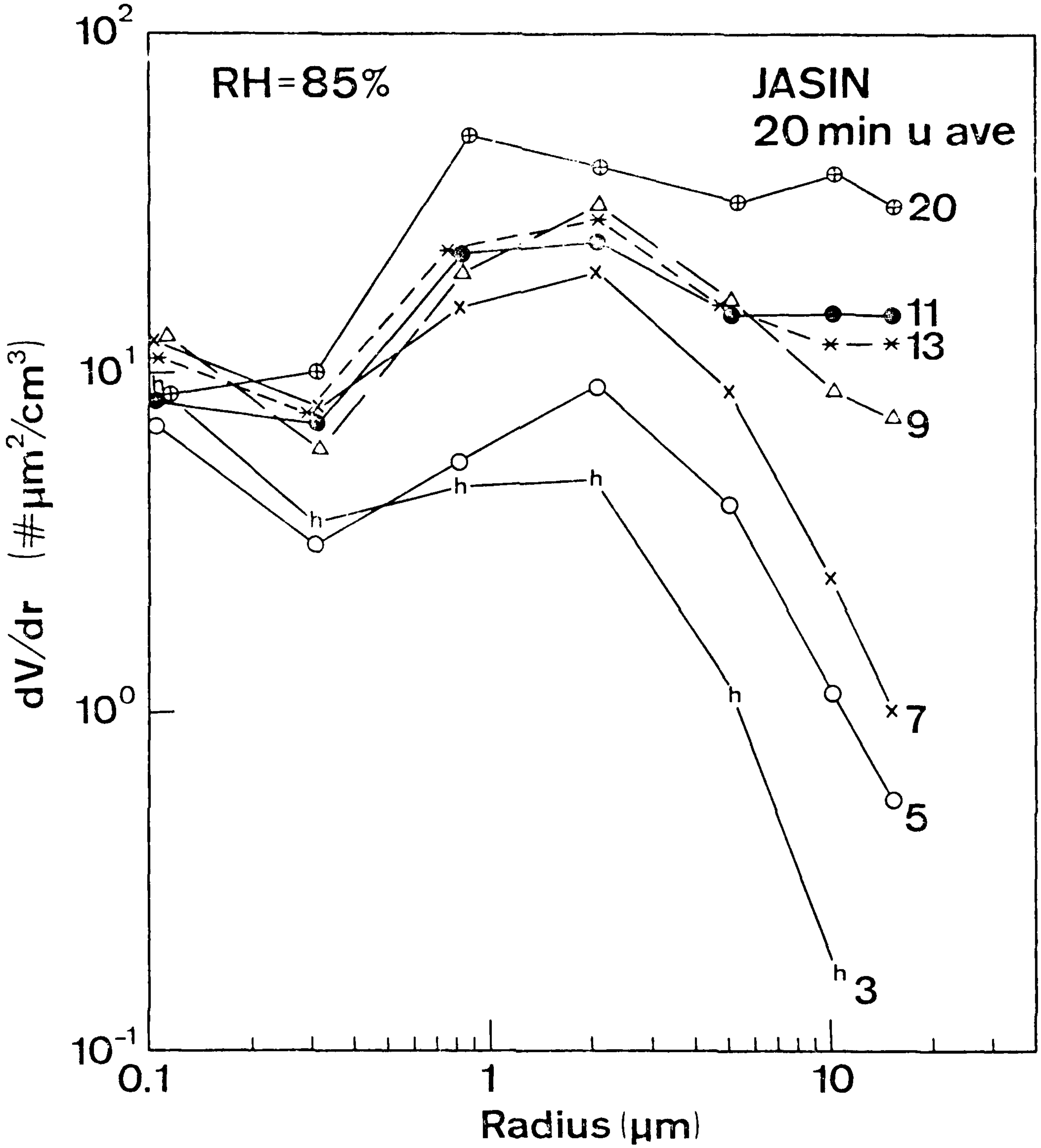

Figure 1. Ensemble average total aerosol volume spectra from JASIN. The number to the right of the spectrum is the wind speed category in meter/second.

where h is the height of the boundary layer, W_e the entrainment velocity and W_s is the Stokes velocity:

$$W_s = 1.57 \times 10^{-2} \left(1 + 1.2 \left(\frac{0.81}{g(s)}\right)^3\right) r^2 \, g^2(s) \qquad (8)$$

where r is in μm and W_s is cm/sec. In this equation we have left out the cloud formation removal mechanism because it acts on a much longer time scale.

C. FLUX SPECTRUM ANALYSIS

The CEWCOM-78 data have been used to evaluate the surface flux term of Equation 7. Since the other terms of the expression were measured, one simply calculates F_s using a rearrangement of the terms.

A period was chosen from CEWCOM-78 where all data were available, the synoptic conditions were fairly stable, the wind speed was reasonably constant and a good mixed layer was present (Table I). There was a slowly decreasing continental influence indicative of steady NW winds (A' in Table I and Figure 2). Subsidence was assumed to be negligible so that W_e = dh/dt. Thus, the final expression used is:

$$F_s = h\frac{dVs}{dt} + \left(\frac{dh}{dt} - W_s\right) V_s \qquad (9)$$

The scatter and uncertainty of the aerosol and mixed layer depth measurements introduces certain trade-offs between averaging times and statistical validity. For short averaging times, the random scatter of the individual terms is considerable leading to large variations in the time derivative terms. By increasing the averaging time to four hours, the F_s "signal-to-noise" becomes more reasonable. A time series of the individual terms of Equation 9 is shown in Figure 3 for two different particle sizes. The average surface production flux spectrum is shown in Figure 4. This flux spectrum applies to the average surface conditions for the entire 20 hour period (average wind speed was about 8 m/sec).

TABLE I. CEWCOM-78 meteorological and aerosol data for the time period analyzed for this report. The aerosol volume spectra density V(R) is given at each radius, R, in μm.

DATE	TIME	U (m/s)	RH (%)	Zi (m)	A	R=.3	R=.8	R=2	R=5	R=10	R=15
								($\mu m^2/cm^3$)			
5/20	1130	7.4	89.4	420	7.8	1.51E+01	1.25E+01	9.79E+00	2.26E+00	8.01E-01	4.37E-01
5/20	1230	7.7	87.6	430	5.2	9.15E+00	1.25E+01	1.19E+01	2.08E+00	6.44E-01	3.27E-01
5/20	1330	8.2	86.4	430	4.5	8.39E+00	1.19E+01	1.22E+01	2.91E+00	1.11E+00	6.28E-01
5/20	1430	8.1	82.1	415	4.7	8.54E+00	1.06E+01	1.14E+01	2.24E+00	7.80E-01	4.20E-01
5/20	1530	8.3	80.9	437	4.0	7.96E+00	1.20E+01	1.12E+01	1.26E+00	2.96E-01	1.27E-01
5/20	1630	8.2	81.1	437	4.0	7.85E+00	1.19E+01	1.36E+01	2.47E+00	7.95E-01	4.11E-01
5/20	1730	9.0	82.5	443	3.6	7.26E+00	1.29E+01	1.34E+01	2.55E+00	8.54E-01	4.55E-01
5/20	1830	10.0	83.8	430	2.6	6.54E+00	1.40E+01	1.32E+01	3.30E+00	1.37E+00	8.19E-01
5/20	1930	10.3	84.3	420	2.4	6.07E+00	1.38E+01	1.32E+01	3.71E+00	1.73E+00	1.11E+00
5/20	2030	9.8	86.8	360	2.1	6.54E+00	1.59E+01	1.42E+01	4.02E+00	1.63E+00	9.58E-01
5/20	2130	10.1	87.7	377	1.9	6.07E+00	1.49E+01	1.32E+01	3.34E+00	1.22E+00	6.78E-01
5/20	2230	9.5	87.6	397	2.7	6.82E+00	1.36E+01	1.28E+01	3.52E+00	1.42E+00	8.33E-01
5/21	2330	9.2	87.6	400	2.7	6.15E+00	1.29E+01	1.27E+01	3.46E+00	1.46E+00	8.91E-01
5/21	30	9.6	87.3	420	3.0	6.14E+00	1.31E+01	1.18E+01	2.58E+00	9.93E-01	5.69E-01
5/21	130	9.2	88.3	490	2.0	5.69E+00	1.35E+01	1.29E+01	3.33E+00	1.26E+00	7.10E-01
5/21	230	8.8	87.4	520	1.7	5.29E+00	1.33E+01	1.34E+01	3.44E+00	1.39E+00	8.18E-01
5/21	330	8.2	86.1	560	2.7	6.11E+00	1.18E+01	1.09E+01	2.18E+00	7.31E-01	3.90E-01
5/21	430	8.5	83.1	597	2.2	5.55E+00	1.16E+01	9.71E+00	1.78E+00	6.59E-01	3.69E-01
5/21	530	8.2	81.3	620	2.0	5.33E+00	1.09E+01	9.75E+00	1.85E+00	7.32E-01	4.31E-01
5/21	630	7.9	79.7	680	1.8	5.02E+00	1.11E+01	1.07E+01	1.96E+00	6.78E-01	3.65E-01

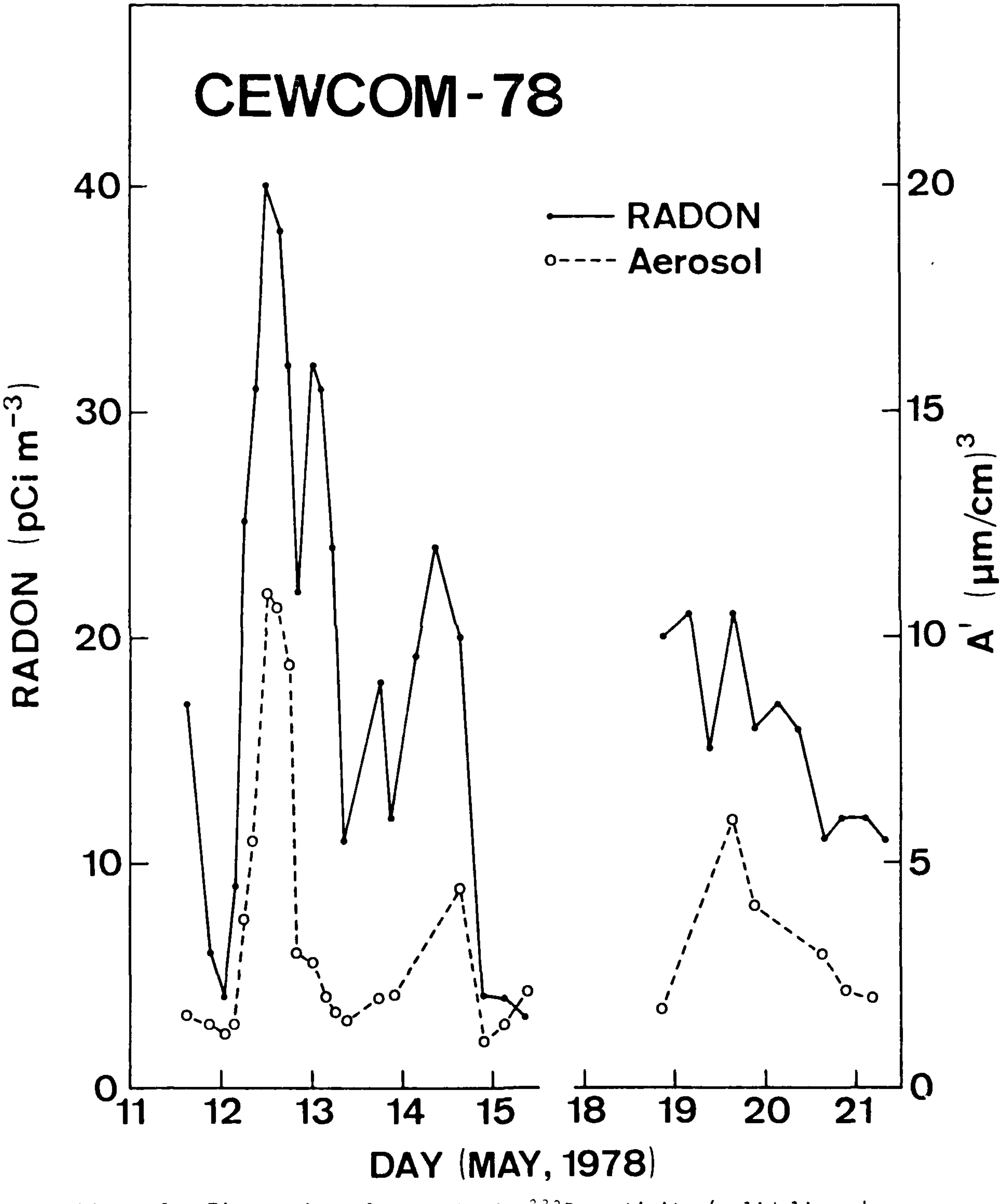

Figure 2. Time series of atmospheric ^{222}Rn activity (solid line, Larsen, Kasemir and Bressan, 1979) and continental aerosol coefficient (dashed line), A'.

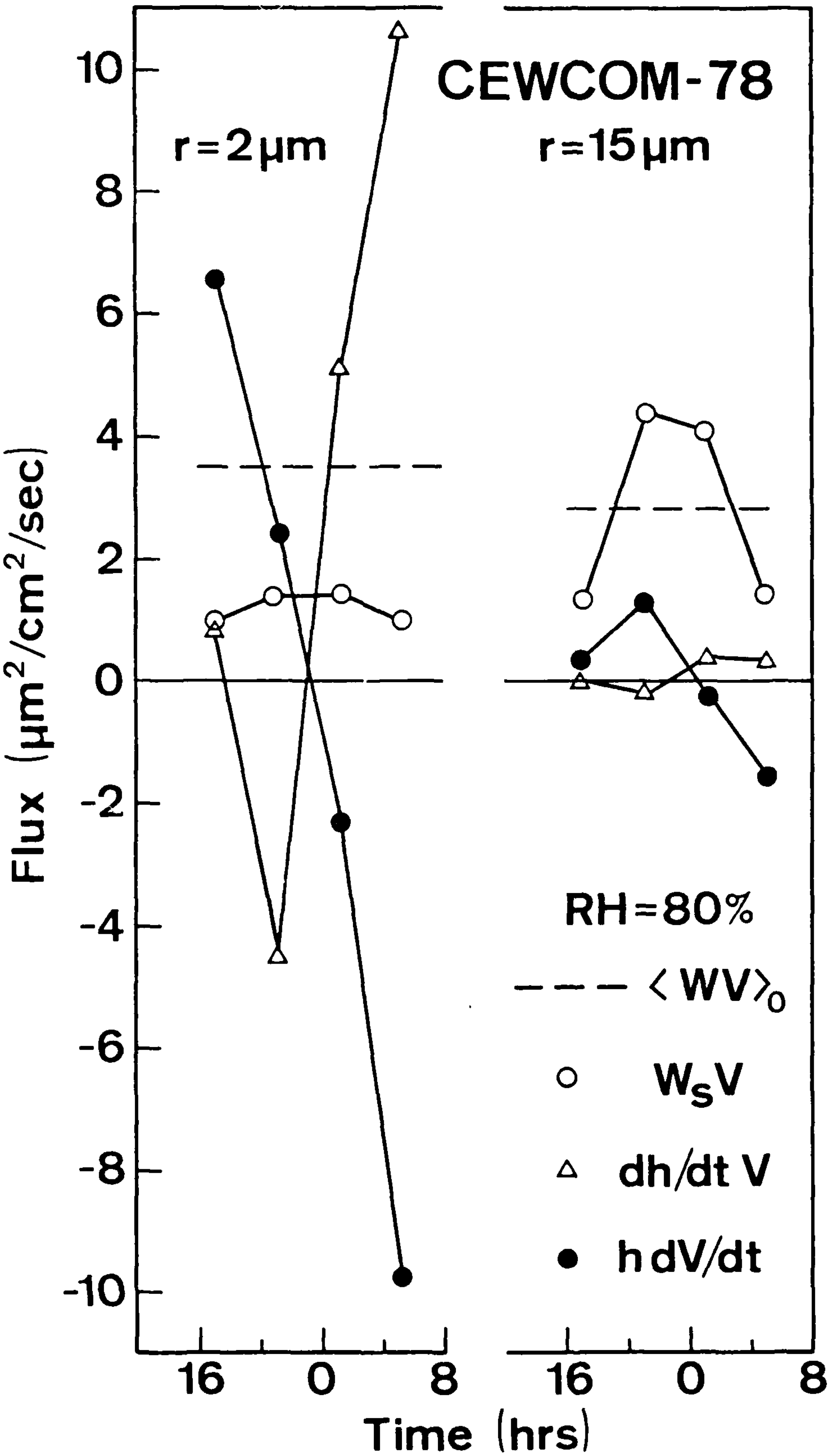

Figure 3. Time series of the terms of Equation 9 for 2 μm radius and 15 μm radius particles. The dashed line is the average contribution of the surface production term and should be the sum of the other three terms.

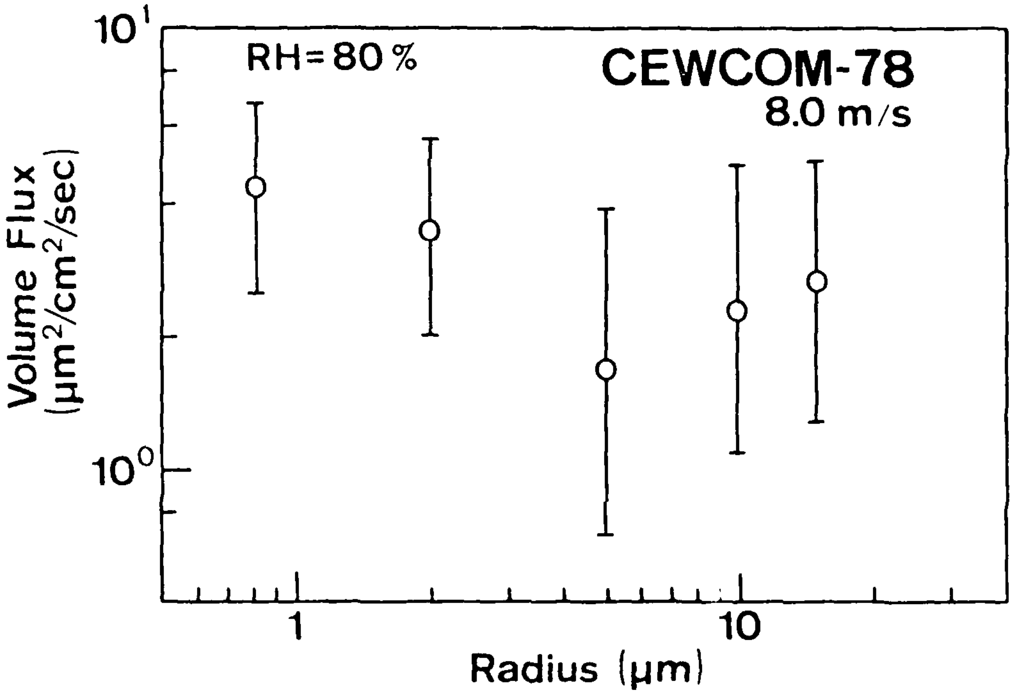

Figure 4. The ensemble average surface flux spectru, $F_s(r)$, for the CEWCOM-78 analysis period (average wind speed, 8 m/sec).

The relative contributions of entrainment, graviational fallout and surface production are nicely illustrated by defining an equivalent surface production vertical velocity, $W_F = F_s/V_s$, so that in equilibrium (dVs/dt = 0)

$$W_F = We + Ws \qquad (10)$$

For the average conditions found in the analysis period, the entrainment and Stokes terms were roughly equal at a particle radius of 5 μm (Figure 5).

D. FLUX SPECTRUM AND WIND SPEED

Because of the reasonably constant wind speeds during the 20 hour CEWCOM-78 period, we were able to improve the statistical certainty for the F_s calculation by combining all the data from the period, assuming that the flux would be a reasonable representation for U = 8 m/sec wind speed. Thus, we now have the surface flux spectrum at a single wind speed. In order to estimate the flux at other wind speeds, we note that the right hand side of Equation 10 is nearly independent of wind speed for equilibrium conditions. Therefore, the flux at one wind speed can be related to the flux at other wind speeds if the equilibrium volume spectra are known.

$$F_s(U_1) = F_s(U_2) \frac{V_s(U_1)}{V_s(U_2)} \qquad (11)$$

We have available from JASIN a large set of ensemble averages of aerosol volume spectra at different wind speeds (Figure 6 and Table II).

It is a simple matter to apply this data to Equation 11, using the CEWCOM-78 aerosol flux and equilibrium spectrum to generate the surface volume flux spectra as a function of wind speed (Figure 7 and Table III).

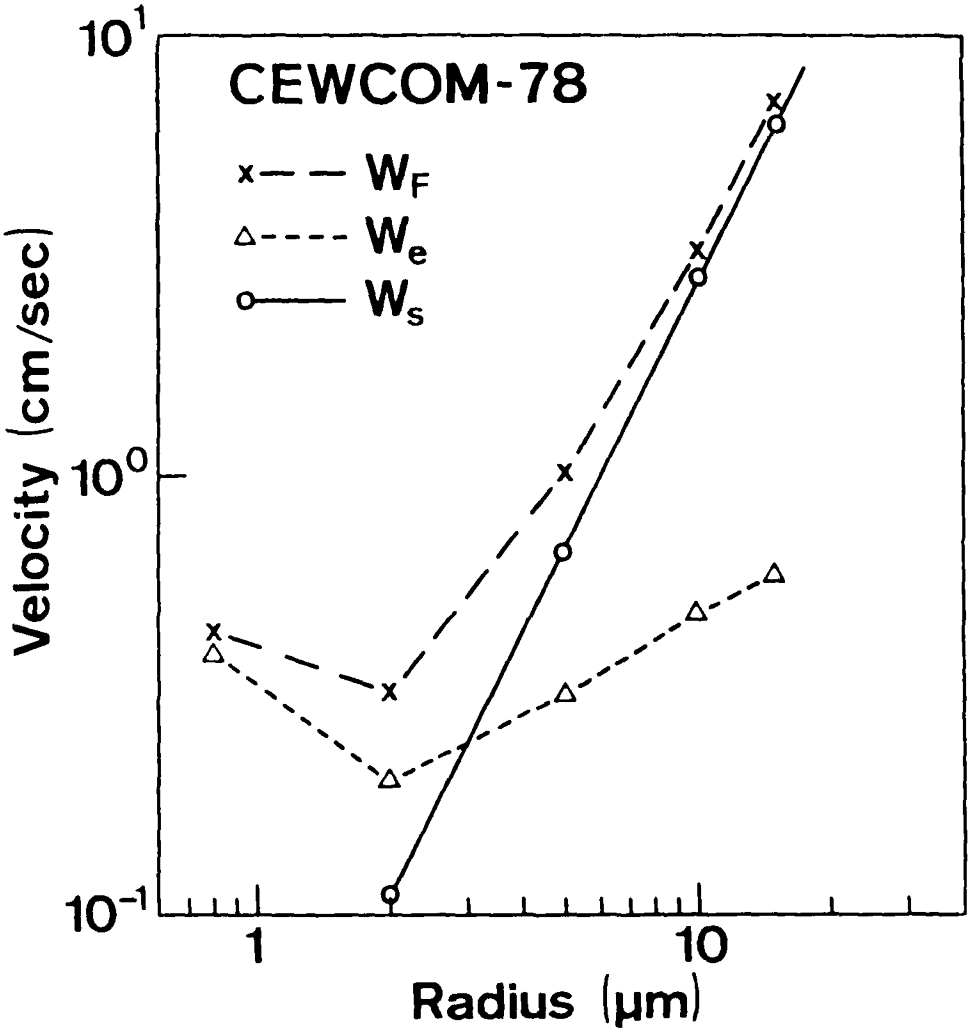

Figure 5. Ensemble average contribution of entrainment (W_e), Stokes fallout (W_s) and surface flux $W_F = F_s/V_s$ assuming a state of dynamic equilibrium.

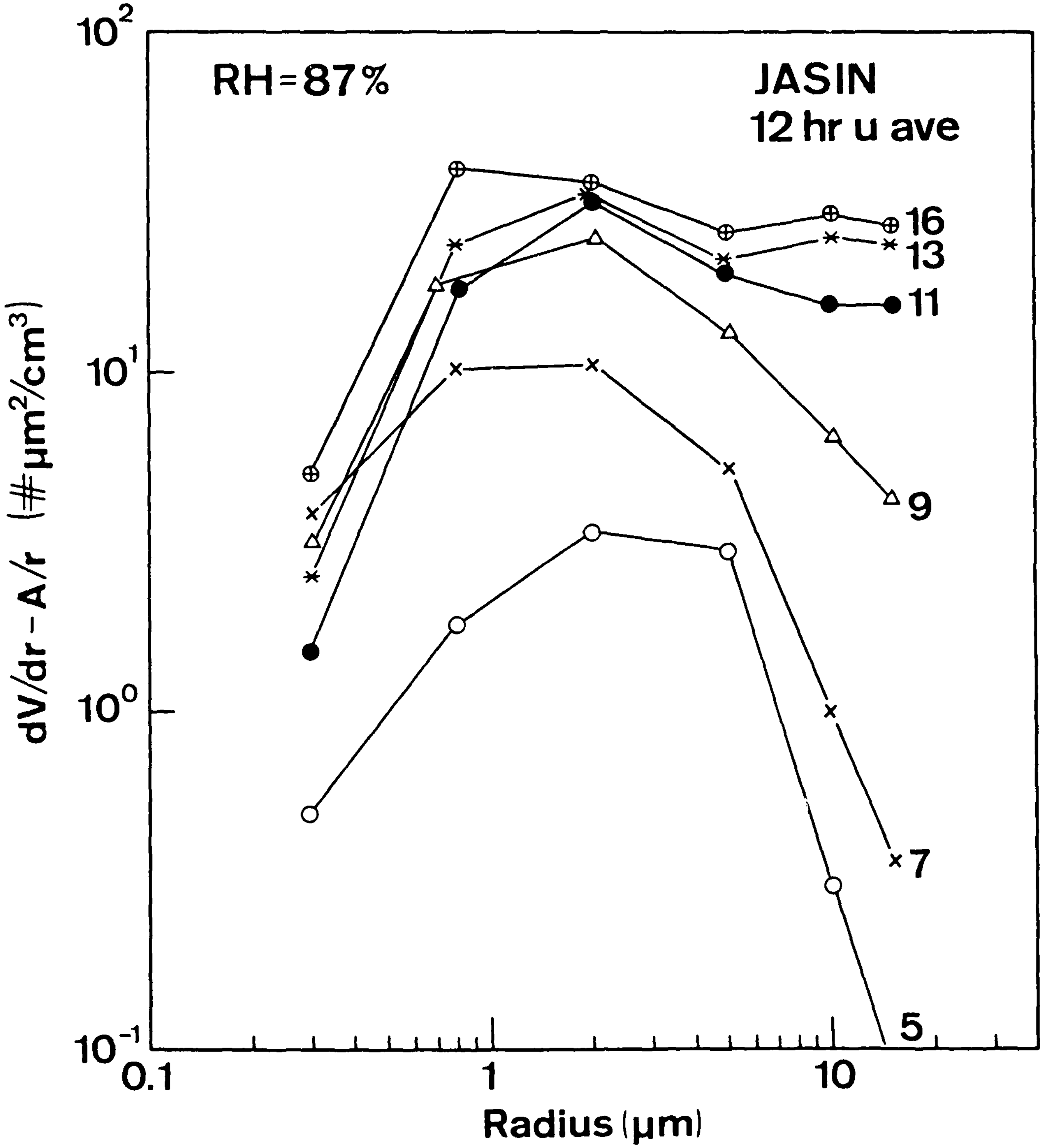

Figure 6. Ensemble average equilibrium sea salt volume spectra from JASIN. The number to the right of the spectrum is the wind speed category in meters/second.

TABLE II. Equilibrium sea-salt aerosol-spectra in $\mu m^2/cm^3$ as a function of radius (μm) and wind speed (m/sec).

u ↓ \ r →	0.8	2	5	10	15
5	2.8	3.4	3	0.3	0.05
7	10	10	5	1	0.35
9	18	25	13	6.4	4.2
11	20	30	20	15	15
13	24	33	22	22	22
16	28	35	28	28	28
8*	9	11	2	0.2	0.4

* CEWCOM-78 Analysis Period

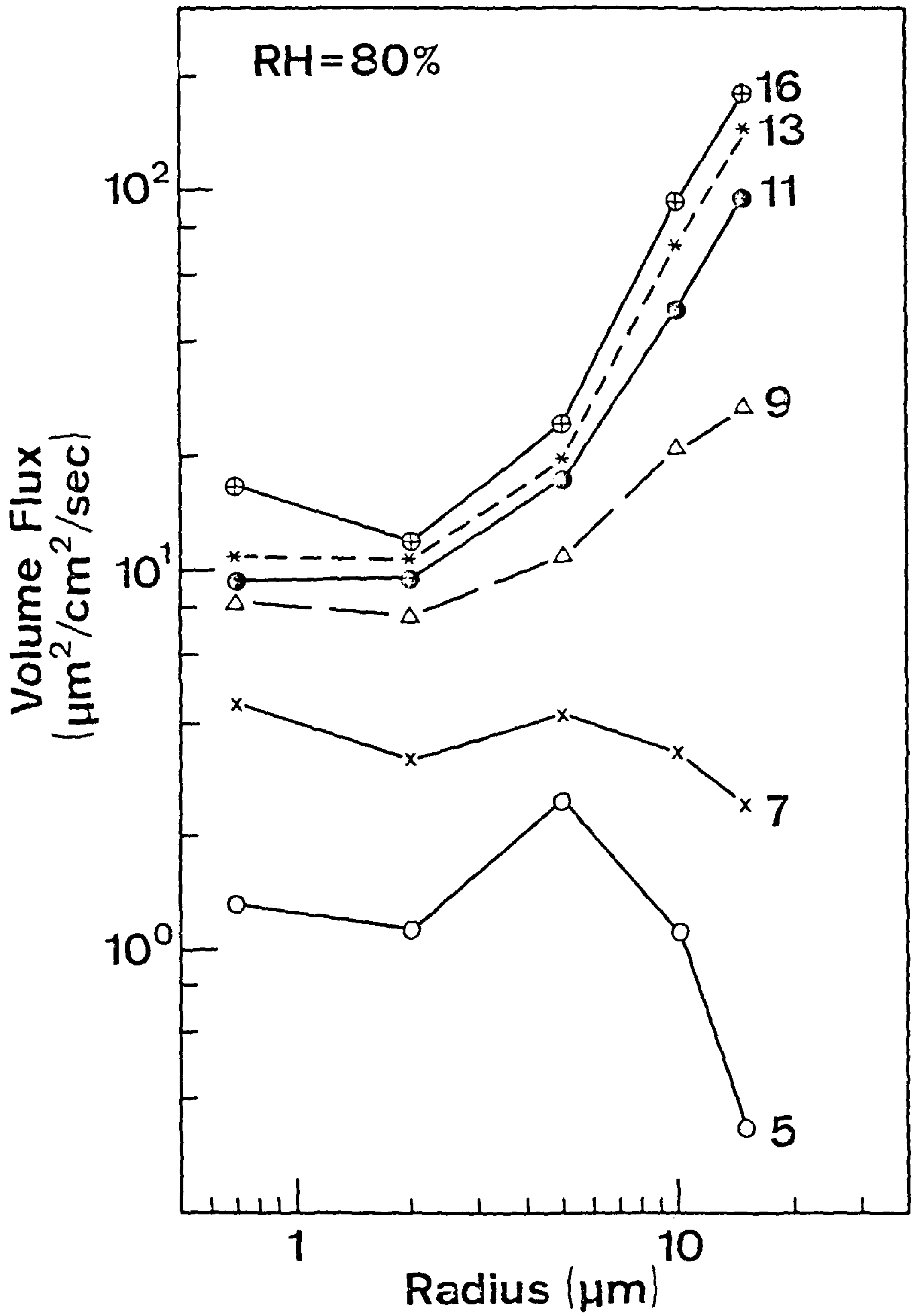

Figure 7. Ensemble average surface flux spectra deduced from Equation 11 and Table II. The number to the right of the spectrum is the wind speed in meters/second.

TABLE III. Surface sea-salt aerosol flux, $F_s(r)$, ($\mu m^2/cm^2/sec$) as a function of particle radius (μm) and wind speed (m/sec).

u \ r →	0.8	2	5	10	15
5	1.3	1.1	2.5	1.0	0.33
7	4.5	3.1	4.2	3.3	2.3
9	8.2	7.7	11	21	27
11	9.1	9.2	17	49	48
13	11	10	19	72	140
16	17	11	24	92	180
8*	4.1	3.4	1.7	2.3	2.6

* CEWCOM-78 Analysis Period

E. NON-EQUILIBRIUM

For time periods of a few hours, the aerosol spectrum may not be in a state of dynamic equilibrium. If we rewrite Equation 7 in the following form:

$$\frac{dVs}{dt} + \frac{1}{\tau_p} = \frac{Fs}{h} \qquad (12)$$

where the time constant, τ_p, is

$$\tau_p = h/(We + Ws) \qquad (13)$$

then we see the analogy of aerosols and a capacitor charged by an applied "voltage," Fs, through a "resistance," h. In this analogy, the "capacitance," is $(We + Ws)^{-1}$.

The response time of the aerosol density is a strong function of particle radius because $Ws \sim r^2$. Values of τ_p for S = 0.8 and h = 400 m are given in Table IV.

TABLE IV. The equilibrium time constant for aerosols at S = 0.8, h = 400 m and We = 0.4 cm/sec.

r, μm	0.5	1	5	10	15
τ_p, hours	28	22	11	3.5	0.5

The boundary layer mixing time, τ_m, which represents the time required for changes in aerosol density to be evenly distributed throughout the marine layer is:

$$\tau_m = h/W_* \qquad (14)$$

For the analysis period ($h = 400$ m and $W_* = 0.6$ m/sec) we find $\tau_m = 0.16$ hours. Therefore, short term variations in mixing volume (dh/dt) will lead to changes in the aerosol density because the production response time is much slower than the mixing time. Thus, for time periods on the order of one hour, changes in the aerosol density (h dVs/dt) will be highly correlated with the mixing volume term (Vs dh/dt).

This effect, which is particularly noticeable for smaller particles (see Figure 3 and Table III) is shown in Figure 8.

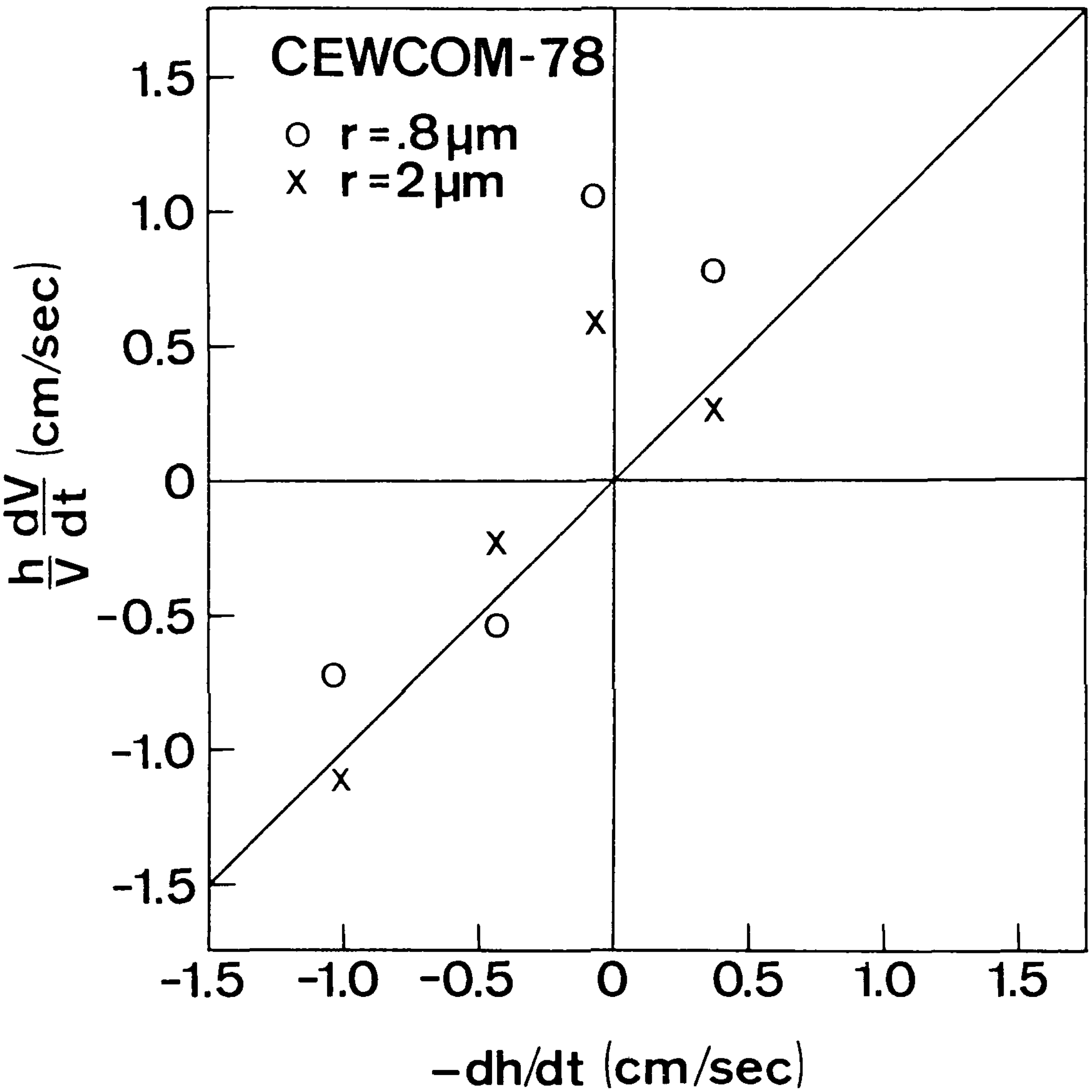

Figure 8. Changes in aerosol spectral density (dV_s/dt) versus changes in mixing volume (-dh/dt). This graph illustrates the dominance of these terms in Equation 9 for short time periods.

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